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TITLE: Size Quantization and Excited States of Associated and Isolated InAs Quantum Dots

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Size quantization and excited states of associated and isolated InAs quantum dots

V. G. Talalaev[†], B. V. Novikov[†], S. Yu. Verbin[†], Dinh Son Thach[†], G. Gobsch[‡],
R. Goldhahn[‡], N. Stein[‡], A. Golombek[‡], J. W. Tomm[§], A. Maassdorf[§],
G. E. Cirlin[¶], V. N. Petrov[¶] and V. M. Ustinov[¶]

[†] Institute of Physics, St Petersburg State University,
198904 St Petersburg, Russia

[‡] Institut für Physik, Technische Universität Ilmenau,
D-98684 Ilmenau, Germany

[§] Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie,
D-12489 Berlin, Germany

[¶] Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Spectra of photoluminescence (PL) including its decay time spectra and spectra of PL excitation (PLE) of heteroepitaxial structures with quantum dots (QD) InAs/GaAs have been studied. Structures had been grown by submonolayer migration-enhanced epitaxy (SMEE) mode on vicinal substrates GaAs at deposited InAs thickness close to critical (1.8 monolayer (ML)). It has been shown that PL structure under study is formed by the recombination emission of different QD families. One family consists of associated QD groups confined by vicinal terraces discretely broadened due to step bunching effect, another family — of isolated QD separated from rest QD array due to wetting layer (WL) ruptures at terrace edges. Excited exciton states of various QD groups have been detected. Their particular features are determined depending on the temperature, power and wavelength of photoexcitation.

Introduction

Additional structure of QD PL spectra is mainly formed by two mechanisms: the excited states recombination and QD size quantization of different origins. However these mechanisms have not been identified. Their evidence in PL spectra of InAs QD grown on vicinal substrates GaAs(100) has been studied here.

1. QD size quantization. Isolated QD

Two-dimensional QD arrays were grown by the SMEE mode on vicinal GaAs substrates at low temperatures (470°C) and growth rates (0.1 ML/s) from InAs layer with thickness close to critical (1.8 ML). For these QD arrays it has been established that at low excitation density (up to 50 W/cm²) the structure of low-temperature PL spectra is formed by QD groups with different sizes. These groups can be attributed to lateral confinement of QD by terraces multiply widened due to step bunching effect on vicinal GaAs surface. Some PL components resulting from such size quantization (A_0 and A_1 bands in Fig. 1) has been observed in PL spectra of relatively high misorientation samples (5 and 7 degrees).

PLE spectra study has determined that A_0 and A_1 QD are associated with WL of InAs (Fig. 2). As the temperature grows up to 200 K the bands get narrower and shift in the long wave direction faster than InAs E_g . It corresponds to the model of thermal redistribution of carriers to larger QD of this group by transfer via WL.

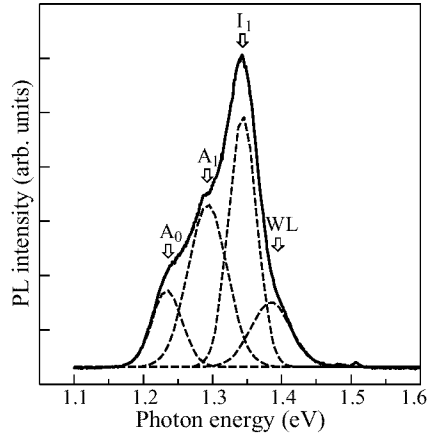


Fig. 1. PL spectrum of InAs/GaAs 7°[001] QD excited by He-Ne laser (20 mW) at $T = 80$ K.

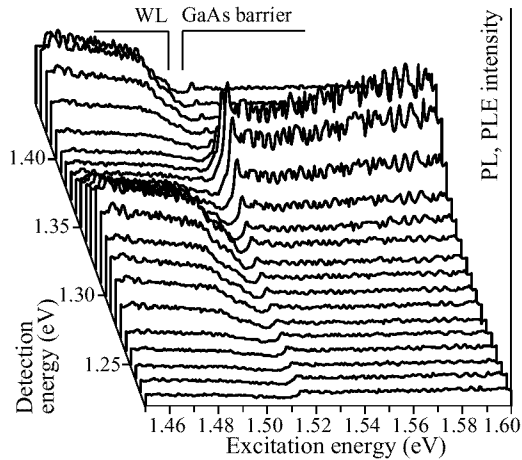


Fig. 2. PLE spectra of InAs/GaAs 5°[010] QD for various detection energy. $T = 5$ K. Excitation by Ti-sapphir-laser (100 mW).

Intensive short-wavelength band I_1 (Fig. 1) of PL spectrum can be attributed to the isolated QD group [1], i.e. nanoislands separated from rest array of QD due to WL ruptures at terrace edges. It has been shown that:

- (1) band I_1 is excited only by carriers from GaAs barrier (Fig. 2);
- (2) its red shift corresponds to the temperature dependence of InAs E_g ;
- (3) as temperature rises up to 200 K the I_1 band broadens in contrast with A-bands [1].

Signs and values of thermal rates of A- and I_1 -bands spectral shifts and halfwidths have been compared in the ranges 5–200 and 200–300 K. It has been concluded that thermal hopping of carriers from shallow isolated QD is not accompanied by their recapture by larger QD. It can be possible if transfer for associated QD takes place via WL and for isolated ones — via GaAs barrier.

The further study has proved that isolated QD are typical not only of vicinal surfaces where they can dominate. WL absence has also been observed at QD growth by SMEE on singular (oriented) GaAs surface. For example, the change of conventional MBE to SMEE

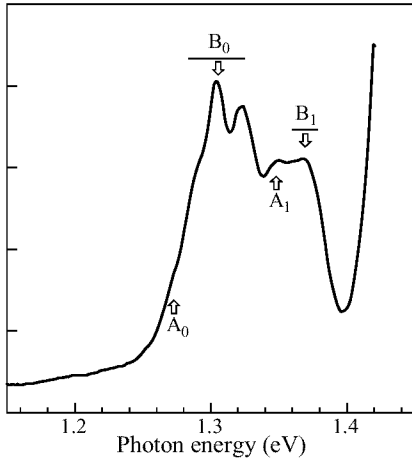


Fig. 3. PL spectrum of InAs/GaAs 7°[001] QD at below-barrier excitation by Ti-Sapphir-laser with 100 mW power and 1.46 eV energy. $T = 5$ K.

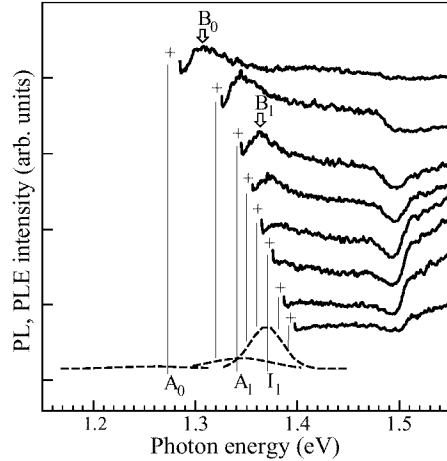


Fig. 4. PLE spectra of InAs/GaAs 7°[001] QD at various detection energies. $T = 5$ K. Excitation by halogen lamp through the monochromator with 20 meV spectral width of slit.

led to the PL band blue shift equal to 55 meV. It corresponds to transition from InAs WL to GaAs barrier.

2. Excited states of associated and isolated QD

A basically different excited state spectral detection of associated and isolated QD has been performed.

Excited states of isolated QD have been observed in PL spectra only at higher (more than 50 W/cm^2) density of above-barrier excitation. Excited state J_1 of isolated QD is higher by 35 meV than ground state I_1 and at high density of excitation (more than 100 kW/cm^2) J_1 dominates in PL spectra of vicinal InAs/GaAs (100) samples with 5°- and 7°-misorientation in [001] and [010] directions.

Excited states of associated QD were observed only at the below-barrier WL excitation (Fig. 3). The discrete inhomogeneity of WL thickness and the corresponding fine structure of B_0 and B_1 excited states have also been observed in this study. For example, in InAs/GaAs 7°[001] sample the state B_0 is higher by 32 meV than ground state A_0 and B_1 — by 20 meV than A_1 (Fig. 3).

It is evident that observed behavior of excited states in associated QD is the result of pyramidal QD wave function localization near the pyramid base [2], i.e. near WL. Then the high density of excitation is not necessary for the observation of B-bands in contrast to J_1 because the density of non-equilibrium carriers in two-dimensional WL is much higher than the surface density of carriers at the excitation of bulk GaAs barrier.

It had been shown earlier [1] that excited states of all QD groups are implicitly evident in the temperature dependence of PL band integral intensity as the deviation of experimental curve from Arrhenius plot. In this way we obtained values of energy gap ΔE between ground and excited states for the InAs/GaAs 7°[001] sample: 30 meV (A_0 – B_0), 26 meV (A_1 – B_1) and 35 meV for I_1 – J_1 .

PLE spectra of this sample are shown in Fig. 4. Observed excitation maxima B_0 and B_1 are shifted against A_0 and A_1 maxima bands by 30 and 23 meV. We have defined B_0

and B_1 peaks as excited states of corresponding associated QD groups.

The parity of energy gaps ΔE in PL and PLE spectra testifies that in QD PL we encounter the radiative transfer between the excited state of one carrier (electron) and the ground state of another one (hole).

Excited state of isolated QD is absent in the PLE spectra (Fig. 4) due to low excitation density. Besides, the energy gap ΔE of isolated QD at $7^\circ[001]$ is equal to LO-phonon energy at InAs-GaAs interface (35 meV [3]). It may be expected that due to the localization of the excited state wave functions at the same interface this resonance leads to a very efficient relaxation of electronic excitation to ground state, i.e. to a very short lifetime of electrons in excited state. The measurement of spectral dependence of PL decay time has shown that at the energy of excited state of isolated QD the fast component of decay curve has evident minimum corresponding to 55 ps.

At the below-barrier excitation the B_0 and B_1 excited state spectrum has fine structure in contrast to the ground state under the same conditions (Fig. 3). It indicates a higher selective sensitivity of excited states to the WL arrangement. The thermal sensitivity of associated QD fine structure and temperature dependence of isolated QD PL in the 5–60 K range is discussed.

Acknowledgments

The work was partly supported by the Russian Foundation for Basic Research (grant 99-02-17780).

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